Oxygen-isotope effect on the in-plane penetration depth in underdoped $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ as revealed by muon-spin rotation

R. Khasanov[(1,2)], A. Shengelaya[(1)], K. Conder[(3)], E. Morenzoni[(2)], I. M. Savić[(4)], and H. Keller[(1)]
[(1)] *Physik-Institut der Universität Zürich, CH-8057 Zürich, Switzerland*
[(2)] *Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland*
[(3)] *Laboratory for Neutron Scattering, ETH Zürich and PSI Villigen, CH-5232 Villigen PSI, Switzerland*
[4] *Faculty of Physics, University of Belgrade, 11001 Belgrade, Yugoslavia*

The oxygen-isotope ($^{16}O/^{18}O$) effect (OIE) on the in-plane penetration depth $\lambda_{ab}(0)$ in underdoped $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ was studied by muon-spin rotation. A pronounced OIE on $\lambda_{ab}^{-2}(0)$ was observed with a relative isotope shift of $\Delta\lambda_{ab}^{-2}/\lambda_{ab}^{-2} = -5(2)\%$ for $x = 0.3$ and -9(2)% for $x = 0.4$. It arises mainly from the oxygen-mass dependence of the in-plane effective mass m^*_{ab}. The OIE exponents of T_c and of $\lambda_{ab}^{-2}(0)$ exhibit a relation that appears to be generic for cuprate superconductors.

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The pairing mechanism responsible for high-temperature superconductivity remains elusive in spite of the fact that many models have been proposed since its discovery. A fundamental question is whether lattice effects are relevant for the occurrence of high-temperature superconductivity. In order to clarify this point a large number of isotope-effect studies were performed since 1987 [1]. The first oxygen-isotope effect (OIE) studies on the transition temperature T_c were performed on optimally doped samples, showing no significant isotope shift [2]. However, later experiments revealed a small but finite dependence of T_c on the oxygen-isotope mass M_O [3, 4, 5, 6], as well as on the copper-isotope mass M_{Cu} [7, 8]. Moreover, a general trend in the dependence of the OIE exponent $\alpha_O = -d\ln T_c/d\ln M_O$ on the doping level was found which appears to be generic for all cuprate superconductors [1, 5, 8, 9, 10]: In the underdoped region α_O is large, even exceeding the conventional BCS-value $\alpha = 0.5$ and becomes small in the optimally doped and overdoped regime.

There is increasing evidence that a strong electron-phonon coupling is present in cuprate superconductors, which may lead to the formation of polarons (bare charge carriers accompanied by local lattice distortions) [11, 12]. One way to test this hypothesis is to demonstrate that the effective mass of the supercarriers m^* depends on the mass M of the lattice atoms. This is in contrast to conventional BCS superconductors, where only the 'bare' charge carriers condense into Cooper pairs, and m^* is essentially independent of M. For cuprate superconductors (clean limit) the in-plane penetration depth λ_{ab} is simply given by $\lambda_{ab}^{-2}(0) \propto n_s/m^*_{ab}$, where n_s is the superconducting charge carrier density, and m^*_{ab} is the in-plane effective mass of the superconducting charge carriers. This implies that the OIE on λ_{ab} is due to a shift in n_s and/or m^*_{ab}:

$$\Delta\lambda_{ab}^{-2}(0)/\lambda_{ab}^{-2}(0) = \Delta n_s/n_s - \Delta m^*_{ab}/m^*_{ab}. \qquad (1)$$

Therefore a possible mass dependence of m^*_{ab} can be tested by investigating the isotope effect on λ_{ab}, provided that the contribution of n_s to the total isotope shift is known.

Previous OIE studies of the penetration depth in $YBa_2Cu_3O_{7-\delta}$ [13], $La_{2-x}Sr_xCuO_4$ [10, 14, 15], and $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_{10+\delta}$ [16] indeed showed a pronounced oxygen-mass dependence on the supercarrier mass. However, in all these experiments the penetration depth was determined indirectly from the onset of magnetization [13, 16], from the Meissner fraction [10, 14], and from magnetic torque measurements [15]. The muon-spin rotation (μSR) technique is a direct and accurate method to determine the penetration depth λ in type II superconductors. In this Letter, we report μSR measurements of in-plane penetration depth λ_{ab} in underdoped $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ ($x = 0.3$ and 0.4) with two different oxygen isotopes (^{16}O and ^{18}O). A large OIE on λ_{ab} was observed which mainly arises from the oxygen-mass dependence of m^*_{ab} .

Polycrystalline samples of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ ($x = 0.3$ and $x = 0.4$) were prepared by standard solid state reaction [17]. Oxygen isotope exchange was performed during heating the samples in $^{18}O_2$ gas. In order to ensure the same thermal history of the substituted (^{18}O) and not substituted (^{16}O) sample, two experiments (in $^{16}O_2$ and $^{18}O_2$) were always performed simultaneously. The exchange and back exchange processes were carried out at 600^{o}C during 25 h, and then the samples were slowly cooled (20^{o}C/h) in order to oxidize them completely. The ^{18}O content in the samples, as determined from a change of the sample weight after the isotope exchange, was found to be 78(2)% for both samples. The total oxygen content of the samples was deter-

mined using high-accuracy volumetric analysis [17]. To examine the quality of the samples low-field (1mT, field-cooled) SQUID magnetization measurements were performed (see Fig. 1). For both concentrations the T_c onset for the ^{16}O samples was higher than for ^{18}O with nearly the same transition width. An oxygen back exchange of the ^{18}O sample ($x = 0.4$) resulted within error in almost the same magnetization curve as for the ^{16}O sample, confirming that the back exchange is almost complete. The results of the OIE on T_c are summarized in Table I. Taking into account an isotope exchange of 78%, we found $\alpha_{\rm O} = 0.22(4)$ for $x = 0.3$ and $\alpha_{\rm O} = 0.37(5)$ for $x = 0.4$, in agreement with previous results [9, 18].

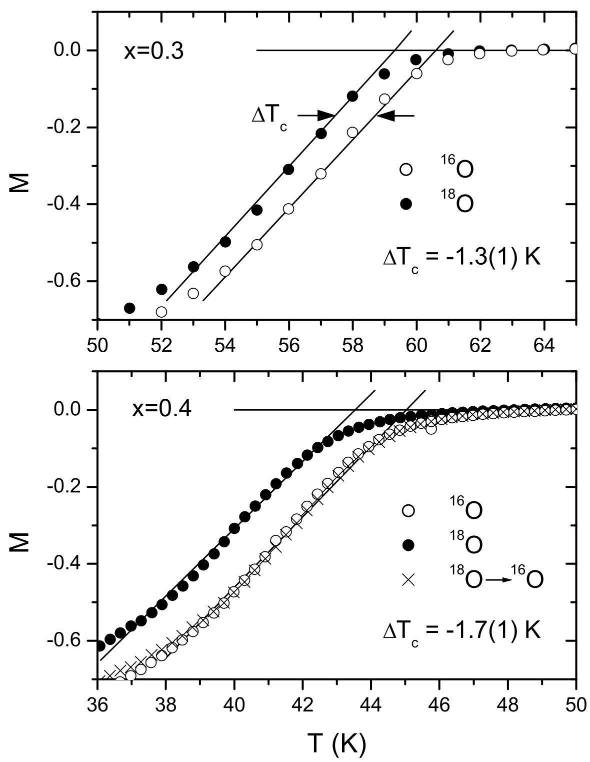

FIG. 1: Section near T$_c$ of the low-field (1mT, field-cooled) magnetization curves (normalized to the value at 10K) for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ (x = 0.3 and 0.4).

The μSR experiments were performed at the Paul Scherrer Institute (PSI), Switzerland, using the πM3 μSR facility. The samples consisted of sintered pellets (12 mm in diameter, 3 mm thick) which were mounted on a Fe_2O_3 sample holder in order to reduce the background from muons not stopping in the sample. The polycrystalline $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples were cooled from far above T_c in a magnetic field of 200 mT perpendicular to the sample disk. Time-differential μSR spectroscopy was employed, from which one can deduce the probability distribution of the local magnetic field $p(B)$ of the vortex state by measuring the time evolution of the muon-spin polarization [19]. In a powder sample the magnetic penetration depth λ can be extracted from the muon-spin depolarization rate $\sigma(T) \propto 1/\lambda^2(T)$, which probes the second moment $\langle \Delta B^2 \rangle^{1/2}$ of $p(B)$ in the mixed state [19, 20]. For highly anisotropic layered superconductors (like the cuprate superconductors) λ is mainly determined by the in-plane penetration depth λ_{ab} [20]: $\sigma(T) \propto 1/\lambda_{ab}^2(T) \propto n_s/m^*_{ab}$.

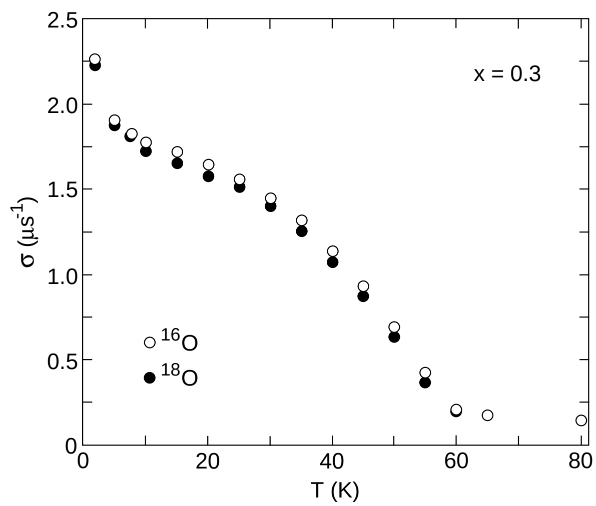

FIG. 2: Temperature dependence of the μSR depolarization rate σ of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ for $x = 0.3$, measured in a field 200 mT (field-cooled).

The depolarization rate σ was extracted from the μSR time spectra using a Gaussian relaxation function $R(t) = \exp[-\sigma^2 t^2/2]$. Figure 2 shows the temperature dependence of the measured σ for the $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples with $x = 0.3$. Similar results were obtained for the samples with $x = 0.4$. It is evident that the values of σ for ^{18}O are systematically lower than those for ^{16}O. As expected for a type II superconductor in the mixed state, σ continuously increases below T_c with decreasing temperature [20]. The sharp increase of σ below $\simeq 10$ K is due to antiferromagnetic ordering of the Cu(2) moments [21]. Above T_c a small temperature independent depolarization rate $\sigma_{nm} \simeq 0.15\ \mu s^{-1}$ is seen, arising from the nuclear magnetic moments of Cu and Pr. Therefore, the total σ is determined by three contributions: a superconducting (σ_{sc}), an antiferromagnetic (σ_{afm}), and a small nuclear magnetic dipole (σ_{nm}) contribution. Because σ_{afm} is only present at low temperatures, data points below 10 K were not considered in the analysis. The superconducting contribution σ_{sc} was then determined by subtracting σ_{nm} measured above T_c from σ. In Fig. 3 we show the temperature dependence of σ_{sc} for the $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples with $x = 0.3$ and 0.4. It is evident that for both concentrations a remarkable oxygen isotope shift on T_c as well as on σ_{sc} is present.

The data in Fig. 3 were fitted to the power law $\sigma_{sc}(T)/\sigma_{sc}(0) = 1-(T/T_c)^n$ [20] with $\sigma_{sc}(0)$ and n as free parameters, and T_c fixed. The values of T_c were taken from the magnetization measurements (see Table I). The

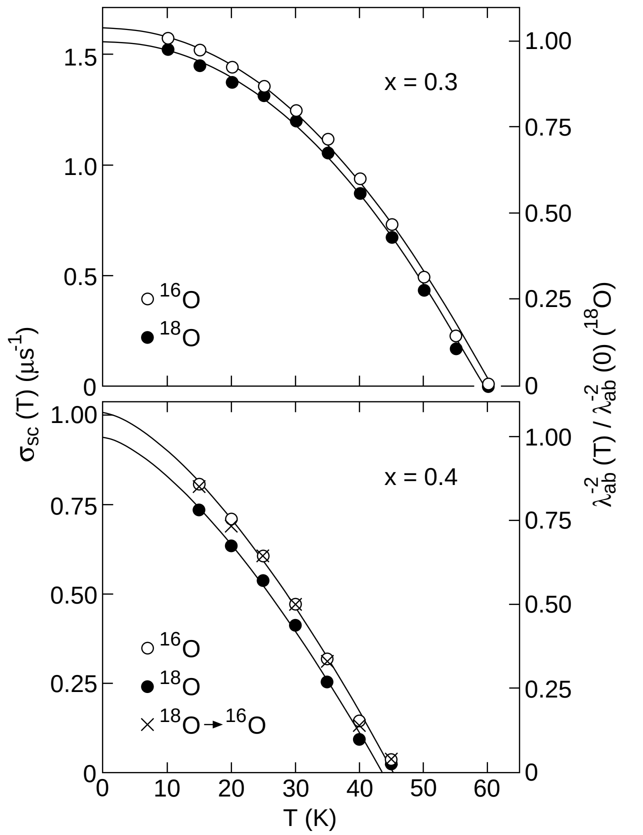

FIG. 3: Temperature dependence of depolarization rate σ_{sc} in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ for $x = 0.3$ and 0.4 (200 mT, field-cooled). On the right axis the normalized in-plane penetration depth $\lambda_{ab}^{-2}(T)/\lambda_{ab}^{-2}(0)(^{18}\mathrm{O})$ is plotted for comparison with Ref. [15]. The solid lines correspond to fits to the power law $\sigma_{sc}(T)/\sigma_{sc}(0) = 1 - (T/T_c)^n$.

values of $\sigma_{sc}(0)$ obtained from the fits are listed in Table I and are in agreement with previous results [21]. The exponent n was found to be $n = 2.0(1)$ for $x = 0.3$ and $n = 1.5(1)$ for $x = 0.4$, which is typical for underdoped YBCO [20]. Moreover, n is within error the same for ^{16}O and ^{18}O. This implies that σ_{sc} has nearly the same temperature dependence for the two isotopes (see Fig. 3). In order to proof that the observed OIE on $\lambda_{ab}(0)$ are intrinsic, the ^{18}O sample with $x = 0.4$ was back exchanged ($^{18}\mathrm{O} \rightarrow {}^{16}\mathrm{O}$). As seen in Fig. 3, the data points of this sample (cross symbols) indeed coincide with those of the ^{16}O sample. From the values of $\sigma_{sc}(0)$ listed in Table I the relative isotope shift of the in-plane penetration depth $\Delta\lambda_{ab}^{-2}(0)/\lambda_{ab}^{-2}(0) = [\sigma_{sc}^{18\mathrm{O}}(0) - \sigma_{sc}^{16\mathrm{O}}(0)]/\sigma_{sc}^{16\mathrm{O}}(0)$ was determined. Taking into account an isotope exchange of 78%, one finds $\Delta\lambda_{ab}^{-2}(0)/\lambda_{ab}^{-2}(0) = -5(2)\%$ and -9(2)% for $x = 0.3$ and 0.4, respectively (Table I). For the OIE exponent $\beta_\mathrm{O} = -d\ln\lambda_{ab}^{-2}(0)/d\ln M_\mathrm{O}$, one readily obtains $\beta_\mathrm{O} = 0.38(12)$ for $x = 0.3$ and $\beta_\mathrm{O} = 0.71(14)$ for $x = 0.4$ (Table I). This means that in underdoped $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ the OIE on λ_{ab}^{-2} as well as on T_c increase with increasing Pr doping x (decreasing T_c). This finding is in excellent agreement with the recent magnetic torque measurements on underdoped $La_{2-x}Sr_xCuO_4$ [15].

TABLE I: Summary of the OIE results for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ extracted from the experimental data (see text for an explanation).

	^{16}O		^{18}O				
x	T_c [K]	$\sigma_{sc}(0)$ [μs^{-1}]	T_c [K]	$\sigma_{sc}(0)$ [μs^{-1}]	α_O	$\frac{\Delta\lambda_{ab}^{-2}(0)}{\lambda_{ab}^{-2}(0)}$ [%]	β_O
0.3	60.6(1)	1.63(2)	59.3(1)	1.57(2)	0.22(4)	-5(2)	0.38(12)
0.4	45.3(1)	1.01(2)	43.6(1)	0.94(2)	0.37(5)	-9(2)	0.71(14)
0.4	45.1(1)[a]	1.01(4)[a]					

[a] results for the back-exchange ($^{18}\mathrm{O} \rightarrow {}^{16}\mathrm{O}$) sample

According to Eq. (1) the observed $\Delta\lambda_{ab}^{-2}(0)/\lambda_{ab}^{-2}(0)$ is due to a shift of n_s and/or m_{ab}^*. For $La_{2-x}Sr_xCuO_4$ several independent experiments [10, 14, 15] have shown that the change of n_s during the exchange procedure is negligibly small. In the present work we provide further evidence: (i) The fully oxygenated $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples ($\delta \simeq 0$) were all prepared under identical conditions, either in a $^{16}O_2$ or $^{18}O_2$ atmosphere [17], and the Pr content x did not change. It is very unlikely that n_s changes significantly upon ^{18}O substitution, and after the back-exchange ($^{18}\mathrm{O} \rightarrow {}^{16}\mathrm{O}$) the same results are obtained (see Figs. 1, 3 and Table I). (ii) According to a model [22] that describes the suppression of T_c in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$, the number of supercarriers decreases linearly with increasing x in the range of $0.05 < x < 0.5$, and consequently $\Delta n_s/n_s = -\Delta x/x$. Moreover, for $0.1 < x < 0.5$ the transition temperature T_c decreases linearly with x, with $\Delta T_c/\Delta x \simeq -150$ K/Pr atom [9]. Combining this two relations one obtains: $\Delta T_c \simeq -150 \cdot x \cdot \Delta n_s/n_s$. Assuming that the observed OIE on λ_{ab}^{-2} is only due to a change of n_s ($\Delta m_{ab}^*/m_{ab}^* \simeq 0$), one can estimate the corresponding shift of T_c. For $x = 0.3$ and $x = 0.4$ one finds $\Delta T_c \simeq -1.8(4)$ K and $-4.2(6)$ K, respectively. The experimental values, however, are much lower (see Fig. 1): $\Delta T_c = -1.3(1)$ K ($x = 0.3$) and $\Delta T_c = -1.7(1)$ K ($x = 0.4$). We thus conclude that any change in n_s during the exchange procedure must be small, and that the change of λ_{ab} is mainly due to the OIE on the in-plane effective mass m_{ab}^* with $\Delta m_{ab}^*/m_{ab}^* \simeq 5(2)$ % and 9(2) % for $x = 0.3$ and $x = 0.4$, respectively. This implies that the effective supercarrier mass m_{ab}^* in this cuprate superconductor depends on the oxygen mass of the lattice atoms, which is not expected for a conventional phonon-mediated BSC superconductor.

In Fig. 4 the exponent β_O versus the exponent α_O for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ is plotted. For comparison we also included the recent magnetic torque results of underdoped $La_{2-x}Sr_xCuO_4$ [15]. It is evident that these

exponents are linearly correlated: $\beta_{\rm O} = A \cdot \alpha_{\rm O} + B$. A best fit yields $A = 1.8(4)$ and $B = -0.01(12)$, so that $\beta_{\rm O} \simeq A \cdot \alpha_{\rm O}$. This empirical relation appears to be generic for cuprate superconductors. Quantitatively one can understand this behavior in terms of an empirical relation between T_c and the μSR depolarization rate $\sigma_{sc}(0)$ [23, 24]. It was shown [24] that for most families of cuprate superconductors the simple parabolic relation $\overline{T}_c = 2\overline{\sigma}(1-\overline{\sigma}/2)$ describes the experimental data rather well (here $\overline{T}_c = T_c/T_c^m$, $\overline{\sigma} = \sigma_{sc}(0)/\sigma_{sc}^m(0)$, and T_c^m and $\sigma_{sc}^m(0)$ are the transition temperature and depolarization rate of the optimally doped system). Using this parabolic Ansatz, one readily obtains the linear relation between $\beta_{\rm O}$ and $\alpha_{\rm O}$: $\beta_{\rm O}/\alpha_{\rm O} = 1 + 1/2\ [(1-(1-\overline{T}_c)^{1/2})/(1-\overline{T}_c)^{1/2}]$. In the heavily underdoped regime

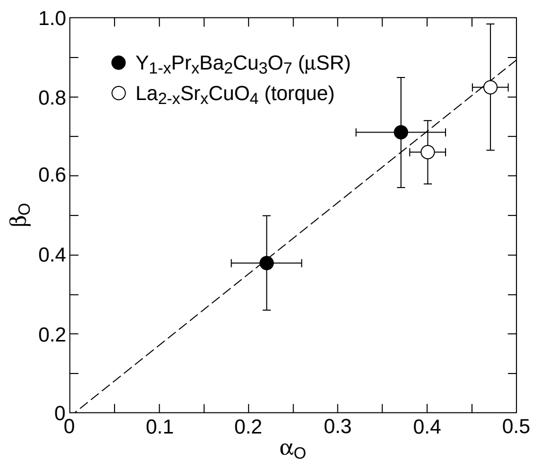

FIG. 4: Plot of the OIE exponents $\beta_{\rm O}$ versus $\alpha_{\rm O}$ for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ ($x = 0.3$ and 0.4) and $La_{2-x}Sr_xCuO_4$ ($x = 0.080$ and 0.086) from [15]. The dashed line represents a best fit to the empirical relation $\beta_{\rm O} = A \cdot \alpha_{\rm O} + B$.

($\overline{T}_c \rightarrow 0$) $\beta_{\rm O}/\alpha_{\rm O} \rightarrow 1$. For the underdoped samples shown in Fig. 4 the reduced critical temperature $\overline{T}_c$ is in the range 0.5 to 0.7, yielding $\beta_{\rm O}/\alpha_{\rm O} = 1.2 - 1.4$, in agreement with $A = 1.8(4)$ obtained from the experimental data. Very recently, it was pointed out [25] that the unusual doping dependence of the OIE on T_c and on $\lambda_{ab}^{-2}(0)$ naturally follows from the doping driven 3D-2D crossover and the 2D quantum superconductor to insulator transition in the underdoped limit. It is predicted that in the underdoped regime $\beta_{\rm O}/\alpha_{\rm O} \rightarrow 1$, which is consistent with the parabolic Ansatz.

In summary, we performed μSR measurements of the in-plane penetration depth λ_{ab} in underdoped $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ ($x = 0.3, 0.4$) for samples with two different oxygen isotopes (^{16}O and ^{18}O). A pronounced OIE on both the transition temperature T_c and $\lambda_{ab}^{-2}(0)$ was observed, which increases with decreasing T_c. The isotope shift on $\lambda_{ab}^{-2}(0)$ is attributed to a shift in the in-plane effective mass m_{ab}^*. For $x = 0.3$ and 0.4 we find $\Delta m_{ab}^*/m_{ab}^* = -5(2)\%$ and -9(2)%, respectively. Furthermore, an empirical relation between the OIE exponents $\beta_{\rm O}$ and $\alpha_{\rm O}$ was found that appears to be generic for various classes of cuprate superconductors. The OIE on m_{ab}^* implies that the superconducting carriers have polaronic character, and that lattice effects play an essential role in the occurrence of high-temperature superconductivity.

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